Estimating Fossil Fuel CO_2, CH_4, and N_2O Emissions Using Tower Measurements in California

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INTRODUCTION

❑ This study reports quantitative emissions assessments for major (fossil fuel CO_2, CH_4, and N_2O) greenhouse gases (GHG) in California

❑ We employ comprehensive measurements and modeling from a collaborative network of tower sites across California, which while focusing on the CalNex intensive, offer a unique opportunity to evaluate seasonal and inter-annual variations in emissions of the sort required for un-biased confirmation of future reductions mandated by AB-32

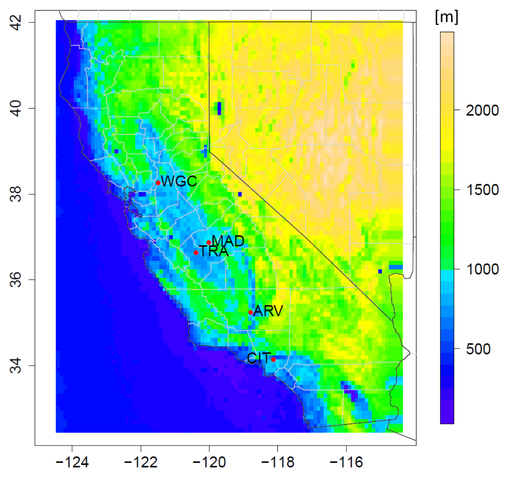
DATA and MODELS

GHG Measurement Network

❑ Fossil fuel CO_2 ($ffCO_2$) was measured at both Walnut Grove (WGC) in spring, 2009 and Pasadena (CIT), during May-June, 2010

❑ CH_4 measurements were made using the CARB-LBNL-NOAA tower network across CA: Arvin (ARV), Madera (MAD), Tranquility (TRA), and WGC

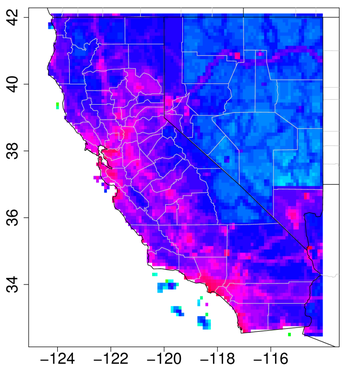
❑ N_2O was measured at WGC for spring 2010



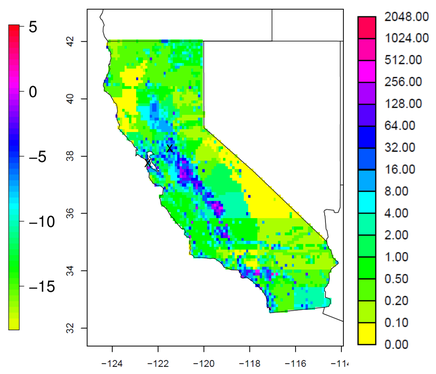
Locations of CARB-LBNL-NOAA GHG Network sites and CIT with predicted monthly mean planetary boundary layer heights (PBLH) for June, 2010, 10:00 PST

DATA and MODELS

Prior $ffCO_2$ and CH_4 Emissions for CA



Vulcan2.0 monthly average $ffCO_2$ emission for June [log(μmol/m²/s)] with a resolution of 0.1 deg.



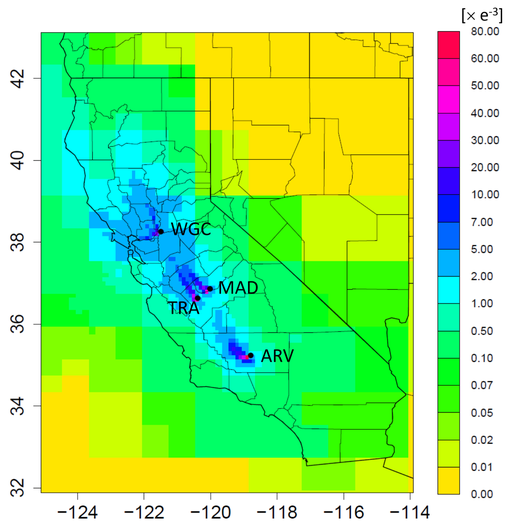
High-resolution (0.1 deg.) CARB-LBNL CH_4 emission inventory [nmol/m²/s]

Footprints

❑ Stochastic Time-Inverted Lagrangian Transport (STILT) model

❑ 500 particles released and transported 5 days backward in time

❑ Mean footprint indicates strong sensitivity of measurements to surface emissions near the tower network sites



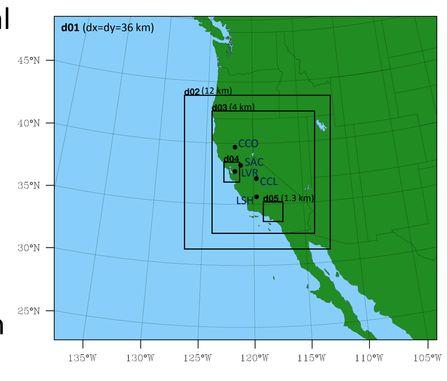
Monthly mean footprint [ppb/(nmol/m²/s)] for September 2010 during the well-mixed period of 1200 – 1700 local (resolution: 0.1 deg.)

WRF Meteorology

❑ North American Regional Reanalysis (NARR) boundary and initial conditions; 6-hour spin-up [Zhao *et al.*, 2009]

❑ Two-way nesting WRF running with four nest levels (five domains)

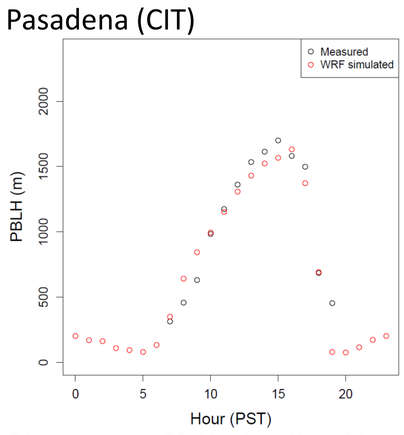
❑ 5-layer thermal diffusion land surface scheme – considers irrigation in CA and performs better than NOAH for PBLH



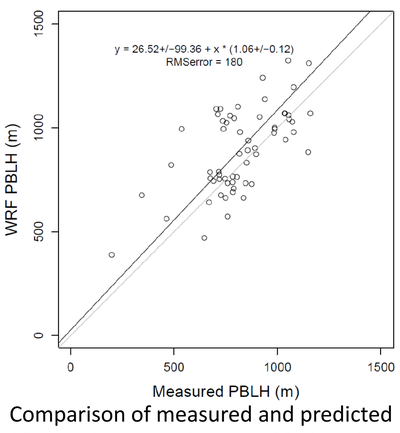
WRF simulation domains, and profiler sites: Chico (CCO), Sacramento (SAC), Livermore (LVR), Chowchilla (CCL), and Lost Hills (LSH)

PBLH Comparison

❑ Predicted PBLH agree well with multi-season daytime radar measurements at five sites in Central CA, and for day-night PBLH measurements by Lidar for May-June 2010 at Pasadena (CIT)



Diurnal cycle of PBLH at Lost Hills, CA for June 2010; measured PBLH available during day time only



Comparison of measured and predicted PBLH at Pasadena for June 2010 during the well-mixed period of 1200 – 1700 local

INVERSION METHODS

Scaling Factor Bayesian Inversion (SFBI)

Model and Solution [Zhao *et al.*, 2009; Tarantola, 1987]

$$y = Hs_p\lambda + v \qquad \hat{\lambda} = \left(K^T R^{-1} K + Q_\lambda^{-1}\right)^{-1}\left(K^T R^{-1} y + Q_\lambda^{-1}\lambda_p\right)$$

y: measured CH_4 – background (marine boundary layer) CH_4
H: sensitivity matrix (footprint)
$\boldsymbol{s_p}$: prior emission
λ: state vector for scaling factor
λ_p: prior for λ $\qquad K=H\,s_p$
R: model data mismatch covariance
$\boldsymbol{Q_\lambda}$: prior covariance matrix for λ
v: error with zero mean and covariance ***R***

Error Covariance

❑ Prior error covariance: diagonal matrix with 50% uncertainty assumption

❑ Model data mismatch covariance: optimized with maximum likelihood estimation

RESULTS

Fossil Fuel CO_2

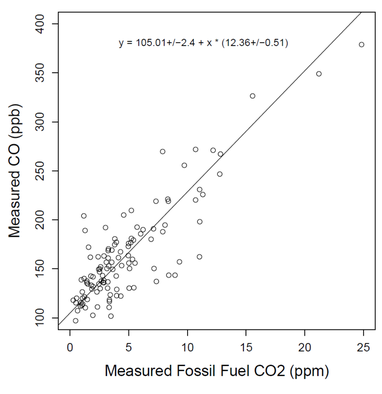
❑ Fossil fuel CO_2 emissions are estimated from radiocarbon $^{14}CO_2$ measured at WGC in spring 2009 and at Pasadena during May - June 2010

❑ 3-hour signals are predicted using Vulcan2.0 emissions during the well-mixed period of 1200 – 1700 local

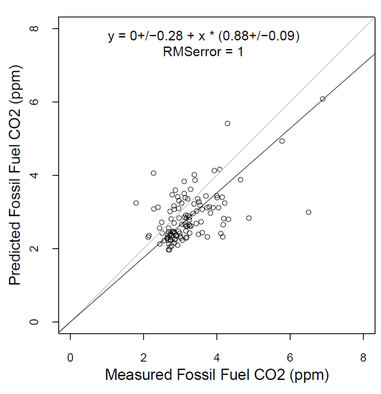
$ffCO_2$ at WGC

❑ Annual (March 2009 – February 2010) $ffCO_2$ (ppm) correlates well with CO (ppb)

❑ $ffCO_2$ emissions are 1.14±0.09x higher than Vulcan2.0 in Central CA



Ratio between measured CO and $ffCO_2$ obtained from individual flask samples



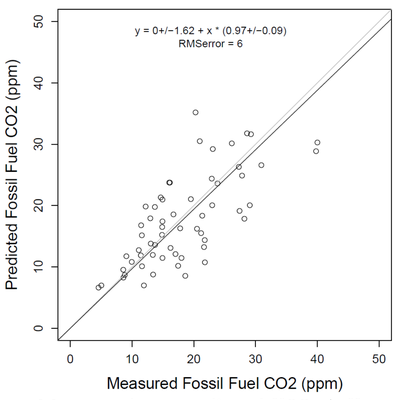
Measured vs. predicted $ffCO_2$ (offset in the regression is adjusted)

$ffCO_2$ at Pasadena

❑ For predictions, nocturnal PBLH in STILT are adjusted based on measured PBLH; nocturnal PBLH adjustment has relatively small effect on day-time signals

❑ Inferred $ffCO_2$ emissions are consistent with inventory within a factor of 1.03±0.09

❑ Results suggest that fossil fuel inventories are accurate at regional scales



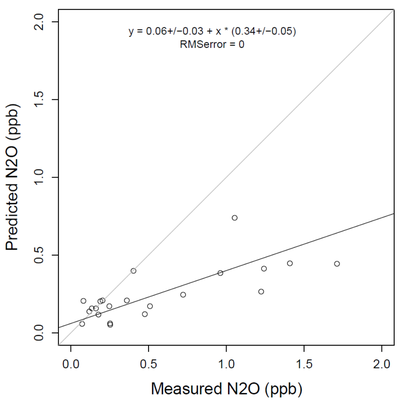
Measured vs. predicted $ffCO_2$ (offset in the regression is adjusted)

N_2O at WGC

❑ N_2O emissions are estimated at WGC for April 2010

❑ 3-hour predictions driven with EDGAR3.2 inventory are compared with 3-hour continuous measurements

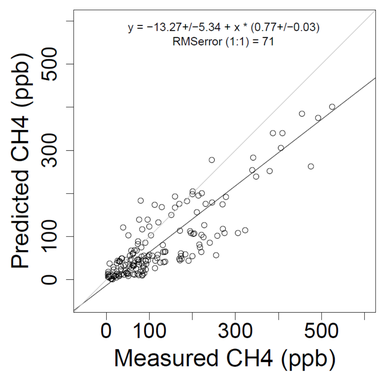
❑ N_2O emissions are 2.94±0.05x higher than EDGAR3.2 for Central CA, consistent with earlier flask-based analyses



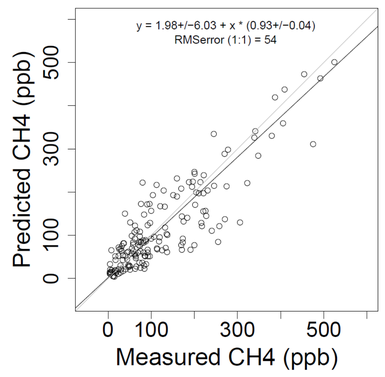
Measured vs. predicted N_2O

CH_4 Inverse Modeling

❑ CH_4 emissions are estimated using a SFBI approach driven by the CARB-LBNL-NOAA tower network measurements and the CARB-LBNL CH_4 emission inventory

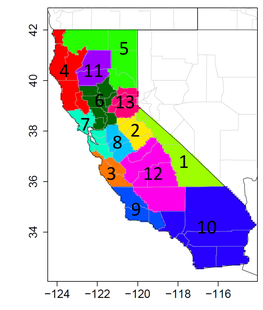


Comparison of 3-hour predicted and measured CH_4 before inversion

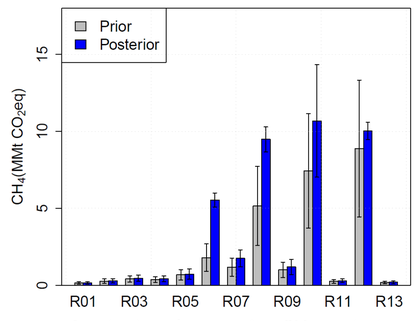


Comparison of 3-hour predicted and measured CH_4 after inversion

❑ Optimized posterior CH_4 emissions for CA sub-regions for September 2010 in the Southern and Northern Central Valley were 1.39±0.09x and 3.09±0.25x larger than the inventory, respectively



Emission sub-region map in CA



A priori and posterior CH_4 emission amount from 13 sub-regions

CONCLUSIONS

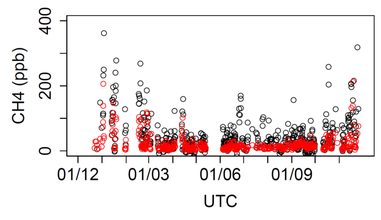
❑ A comprehensive study of GHG emissions including the CalNex campaign is presented using a collaborative network of tower sites across California

❑ $ffCO_2$ simulations at WGC and Pasadena show emissions are consistent with Vulcan2.0 inventory

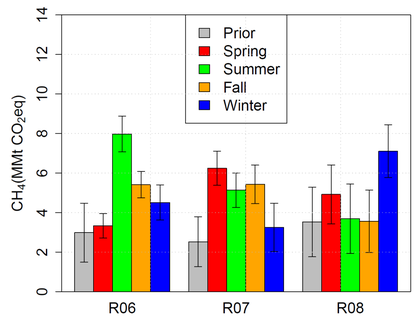
❑ CH_4 inversion driven by the CARB-LBNL-NOAA network effectively constrains emissions in Central Valley

❑ N_2O signals at WGC confirm that actual emissions are significantly higher than EDGAR3.2 estimates

❑ The inversion results driven by the network CH_4 observations are consistent with a longer term seasonal analysis of Walnut Grove CH_4 observations



Long-term seasonal variation (dates given as DD/MM) of 3-hour measured (black) and predicted (red) local signals for well-mixed periods between December 2007 and November 2008



A priori and posterior CH_4 emission amount from three sub-regions near WGC based on a long-term seasonal analysis; Inventory in Zhao *et al.* [2009] is used

Acknowledgments

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References

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- Zhao, C., A. E. Andrews, L. Bianco, J. Eluszkiewicz, A. Hirsch, C. MacDonald, T. Nehrkorn, and M. L. Fischer (2009), Atmospheric inverse estimates of methane emissions from Central California, *J. Geophys. Res.*, 114, D16302, doi:10.1029/2008JD011671.